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A perspective on the state of Deepwater Horizon oil spill related tarball contamination and its impacts on Alabama beaches

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The Deepwater Horizon (DWH) accident spilled over 785 million liters of crude oil into the Gulf of Mexico (GOM). A substantial fraction of the spilled oil impacted the northern GOM shoreline, including Alabama beaches. The beached oil was in the form of brownish-orange, water-in-oil emulsion, commonly known as mousse. Although significant remediation efforts were undertaken to clean the contaminated beaches, oil residues in the form of tarballs continue to contaminate various GOM beaches. This study reviews recent literature related to the DWH tarball contamination problem and its impacts on GOM beaches, primarily focusing on the beaches located in Alabama. Though the DWH oil spill is an unfortunate disaster, for researchers it constitutes a large-scale experiment conducted on a natural system. This anthropogenic experiment has taught scientists numerous useful lessons and has also posed several challenging questions, some of which are discussed in this review.

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Introduction

On April 20, 2010, Deepwater Horizon (DWH), a semisubmersible oil exploration drilling rig, operated by British Petroleum (BP) in the Macondo Prospect 252 (MC252), experienced a catastrophic well blowout resulting in a major explosion. This accident released about 785 million liters of crude oil into the waters of the Gulf of Mexico (GOM) over 87 days, and the well was eventually capped on July 15th, 2010. Several initial assessments were rather optimistic, and some marine chemists even boldly predicted that "*we won't see a 'black tide' in this spill as we did after Exxon Valdez*" [1]. Interestingly, despite all the mitigation efforts, a substantial amount of the spilled oil was transported by ocean currents and was deposited on the beaches located along the northern GOM shores in Florida, Alabama, Mississippi, Louisiana, and Texas. The oil deposition was mostly in the form of a brownish-orange tide (instead of the black tide observed after the Exxon Valdez disaster). The sandy Alabama beaches located in between Orange Beach and Fort Morgan were some of the most highly contaminated amenity beaches by these brown oil tides [2,3]. The presence of oil on these beaches had an enormous impact on the economy of the Gulf Coast of Alabama. For example, Winkler and Gordan [4] estimated that the condominium prices in this region dropped by about 50%, and they also pointed out that macroeconomic studies have estimated the net adverse economic impacts resulted in an output loss of about $1.8 billion.

The brownish oil tides started washing on the shores of Alabama from the first week of June 2010. The oil that impacted the Alabama beaches was predominantly in the form of a highly viscous, neutrally buoyant, brownish, water-in-oil emulsion [5•], commonly known as the 'chocolate mousse.' Over the next few weeks, an unknown quantity of this emulsified oil interacted with nearshore suspended sediments and sank to the sandy bottom forming oil mats (commonly referred to as tarmats). Later these tarmats were exposed and broken apart by coastal processes forming fragments of oil–sand residues (commonly referred to as tarballs). These tarballs continue to contaminate Alabama beaches [6••]. The objective of this article is to review the current state of DWH tarball contamination problems in the northern GOM beaches and their environmental impacts, primarily focusing on studies related to Alabama beaches. Furthermore, we will contrast the optimistic forecasts made by some of the early DWH oil spill studies with the field observations made over the past 10 years to understand the relative importance of various natural processes during the recovery period of large oil spill events.

Background tarball contamination in Alabama beaches before the DWH oil spill

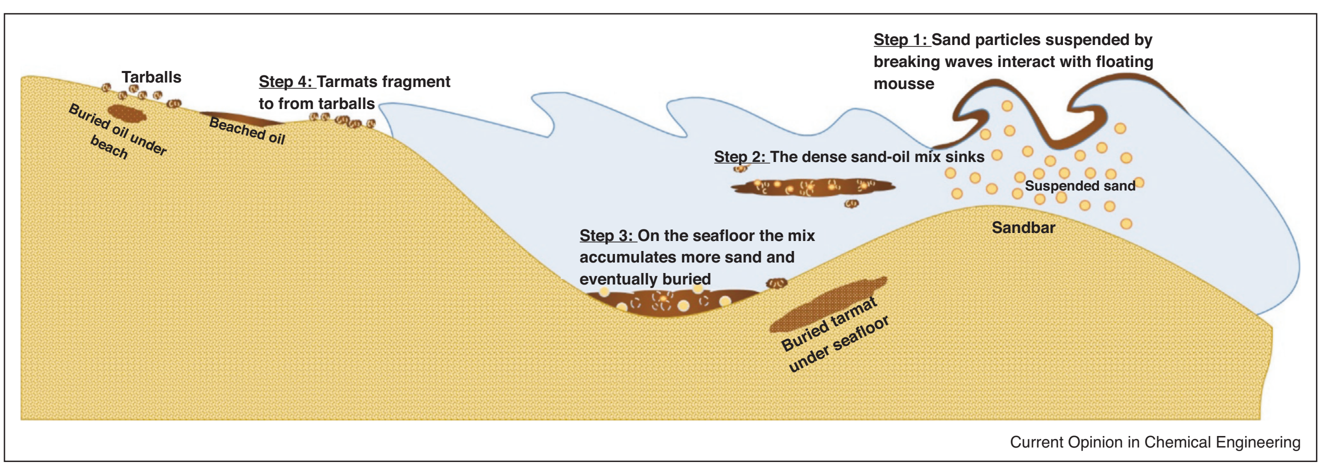
Clement *et al.* [2] completed a study to document tarball contamination deposition patterns along Alabama beaches during various stages of the DWH oil spill event. They also reviewed published studies to estimate the background oiling levels for Alabama beaches. One of the first attempts to evaluate the background oil levels in Alabama was completed by the Unified Area Command (UC) team. This team investigated Alabama beaches in May 2010, a few weeks before the DWH oil started to impact these beaches, and summarized their findings in a research report [7]. They surveyed 8.4 km long sandy beaches in the City of Orange of Beach and found no tarballs. They recovered 40 tarballs with an average size of 0.4 cm when they surveyed another 40.5 km of sandy beaches located from Gulf Shores to Fort Morgan in Alabama. Before this investigation, Romero *et al.* [8] completed a tarball survey in the Florida Panhandle region and concluded that Florida's panhandle beaches (which are located to close to Alabama beaches) are quite pristine since most of them had no observable tarballs. Although the GOM has several natural oil seeps, all known oil seeps are located about 200–300 km away from the shoreline, and currently, there is no published record of any beach contamination problem due to these natural seeps [2]. Based on these background data, the tarball level for Alabama beaches before the DWH oil spill can be estimated to be as 1–2 g of highly weathered tarball residues per kilometer of shoreline per year [2].

Tarball contamination of Alabama beaches after the DWH oil spill

Nixon *et al.* [9] developed a comprehensive database for mapping the GOM shoreline region that was contaminated by the DWH oil spill. They completed several surveys between May 5th, 2010, to March 25th, 2014, across several impacted coastal areas in Florida, Alabama, Mississippi, and Louisiana. Their study concluded that about 69 km of Alabama beaches had heavier/lighter persistent oiling. Clement *et al.* and Hayworth *et al.* [2,3] presented the results of multiple DWH oil spill assessment surveys completed along Alabama beaches extending from Orange Beach to Fort Morgan in the last ten years. During a field survey completed on January 30th, 2016, their team recovered over 200 tarballs with sizes ranging from 1 to 10 cm and a total weight of about 1300 g from Fort Morgan beaches [2]. More recently, Arekhi *et al.* [6••] reported a field survey completed along Fort Morgan beaches on March 17th, 2020, where they observed a highly contaminated area scattered with a large number of tarballs. They completed a detailed survey within a 50 m × 50 m area and recovered over 150 (total number of) tarballs with sizes ranging from 2 cm to 10 cm. The total weight of the tarballs collected from this area was 1250 g.

It is clear from these recent field surveys that tarballs related to the DWH oil spill continue to persist along Alabama beaches. These tarballs primarily originate from DWH oil deposits buried along the GOM shoreline. During the early days of the DWH oil spill event, a substantial portion of the oil that washed along these beaches interacted with suspended sediments and sank [2,10]. These sunken oil–sand aggregates got trapped in the nearshore environment in the form of tarmats and serve as tarball sources. Gustitus and Clement [10] reviewed various fate and transport processes occurring near sandy beach environments to develop a conceptual model for the nearshore oil sinking process that resulted in the formation of these sunken oil deposits (see Figure 1). As shown in the figure, the DWH tarmats were formed from the emulsified brownish oil mousse after it interacted with sand particles through a series of transport steps [2,10,11]. The first transport step occurred either near the subtidal or intertidal zone, where the

Figure 1



A conceptual model for describing the formation of tarmats and tarballs from floating DWH oil spill mousse (modified from Gustitus and Clement [10]).

waves breaking on the sandbars forced the entrained sand particles to interact with the floating oil mousse. After accumulating enough sand, the density of the sand-mousse mixture became higher than that of the water, and the sand-mousse mixture sank. Once the mixture reached the ocean bottom, it accumulated additional sand and formed large mats of oil (dimensions ranging from 1 to 10 m). With time, these tarmats were broken apart into smaller fragments (sizes ranging from 1 to 10 cm). Occasionally, small chunks of the floating mouse also directly interacted with the sand particles to form sunken tarballs. It is important to note that tarmats and tarballs are identical oil–sand aggregates; they are named differently based on their physical dimensions. They are termed tarballs when they are small (typically less than 10 cm), and tarmats when they are large (above a meter). Anything intermediate is often called a tar patty.

Gustitus and Clement [10] proposed a formal nomenclature for classifying oil–sand aggregates of different sizes typically found in the nearshore sandy beach environment after an oil spill. In the current study, however, we will continue to use the popular nomenclature 'tarballs and tarmats' that is routinely used to refer to DWH oil–sand aggregates. Interestingly, large deposits of oil–sand aggregates (such as tarmats) are rarely formed in the natural environment. Several recent oil spills that have occurred close to sandy beaches did not produce any appreciable amount of sunken oil along the shoreline. Our team was involved in field efforts to sample oil spill residues along sandy beaches near Galveston, Texas, after the 2014 Galveston oil spill [12], as well as along Chennai City beaches in India after the 2017 Chennai oil spill [13]. Our field observations show that these two oil spills did not form any sunken tarmats, although both of these spills did deposit a considerable amount of emulsified floating oil on the shoreline. One of the major differences is that the Galveston and Chennai oil spills occurred close to the shoreline and hence had very little time to form thick emulsions. Also, they were substantially smaller spills (in terms of the overall volume of the spill and duration) when compared to the DWH oil spill.

DWH tarballs have several unique physical characteristics. Before the DWH oil spill, the word 'tarball' has been mostly used to refer to highly weathered tarlike black rubbery residues formed from old oil spills. In studies published during the early days of the DWH oil spill, scientists speculated that the DWH spill would result in the formation of highly weathered, non-toxic tarballs. For example, the chemists who examined some of the initial DWH oil spill samples concluded that the residues "*looked like roof tar and such goo is expected to eventually form tarballs. These tarballs are going to be very sticky but not very toxic* [1]." It is unclear what the authors meant by the word 'eventually', but the conventional wisdom is that highly weathered, floating black tarballs are often formed when the spilled oil is weathered by the deep ocean-scale processes for a very long period (several months to years). The DWH tarballs, however, were formed rather rapidly, within days, by nearshore processes. They trapped the relatively fresh brownish oil mousse that contained a significant amount of toxic chemicals, including polycyclic aromatic hydrocarbons (PAHs) [14]. When these oil–sand aggregates are buried in the intertidal zone in large volumes as tarmats, they are difficult to locate and remove. The current state of practice for removing tarmats primarily depends on manual excavation methods, which can be laborious and highly expensive [11].

Methods for identifying DWH tarballs

Since DWH tarballs can potentially be mixed with other types of oil spill residues, researchers have attempted to develop field protocols to differentiate the DWH tarballs from other conventional tarballs. Han and Clement [15] proposed a field-testing protocol for identifying the DWH tarballs purely based on their physical characteristics. Their study found that DWH tarballs are fragile, sticky, brownish, dense objects that contain a considerable amount of sand (about 80% sand with a specific gravity of about two). On the other hand, the traditional tarballs are a rubbery or hard material and are typically non-sticky, black objects that contain very little sand. Most traditional tarballs float on water and hence have a specific gravity value of less than one. Han and Clement's study also conducted chemical fingerprinting of several petroleum biomarker compounds present in different types of tarballs to validate that the fragile, brownish, sticky tarballs were indeed formed from the DWH oil spill.

In the published literature, researchers have used the chemical fingerprints of different types of petroleum biomarker compounds to distinguish the DWH tarballs from other oil spill residues that could have potentially originated from natural seeps, past oil spills, accidental releases from oil exploration, production of crude oil, and petroleum transportation activities [15,16]. Petroleum biomarkers are geochemical organic compounds naturally present in crude oils. Their composition in crude oil is unique and can be related to their biological precursors. Terpanes, steranes, and triaromatic steranes are the most commonly used biomarker compounds used for source identification in oil spill studies [6••,15–17]. Past studies have compared the diagnostic ratios of different types of petroleum biomarkers present in the DWH tarballs and have demonstrated that the DWH crude oil residues have a unique chemical fingerprint [6••,15]. These biomarker diagnostic ratios are also highly stable and can resist various environmental weathering processes. Arekhi *et al.* [6••] investigated the stability of the biomarkers using DWH tarball samples that have weathered in the Alabama coastal environment for over 10 years. They concluded that higher molecular weight terpanes (heavy tricyclic terpanes and all pentacyclic terpanes) and higher molecular weight steranes (diasteranes, ergostanes, and stigmastanes) are stable compounds. Their data also showed that all the homohopanes (in the range of H31–H35) remained stable even after 10 years of natural weathering. Although some lower molecular weight tricyclic terpanes and steranes have shown some degree of weathering, the diagnostic ratios of different pairs of hopanes, steranes, and triaromatic sterane compounds remained stable, and hence these compounds can be reliably used for source identification.

The fate of toxic petroleum hydrocarbons present in DWH oil spill residues

Recent studies have reported that toxic compounds present in submerged DWH oil spill residues tend to weather rather slowly. Bagby *et al.* [18] studied the biodegradation patterns of DWH oil collected at the seafloor at depths ranging from 1029 to 1912 m and found that several toxic PAHs experienced two distinct degradation phases: rapid degradation while oil particles remained suspended, followed by slow degradation after deposition. The extent of biodegradation for any given sample was influenced by the hydrocarbon content, and highly contaminated samples tend to degrade rather slowly. Yin *et al.* [14] analyzed several tarball samples recovered from the subtidal zone of Alabama beaches at different times and found that several PAHs weathered by a large fraction when the oil was floating over the open ocean. The weathering rate of PAHs slowed significantly once the oil was trapped and buried within the coastal environment. Several higher molecular weight PAHs trapped in highly contaminated residues have nearly stopped degrading in the buried oil. John *et al.* [19] extracted the residual oil from DWH tarballs and exposed it to the sunlight. Their results show that upon exposure to sunlight the weathering of hydrocarbons resumed, indicating that photodegradation should have played a significant role in the weathering of hydrocarbons when the oil was floating over the ocean. They hypothesized that the weathering rates of PAHs trapped in buried tarmats and tarballs must have slowed down since the oil was isolated from direct exposure to sunlight. Aeppli *et al.* [20] completed a detailed study to investigate the persistent and bioavailability of oxygenated hydrocarbon (OxHC) compounds formed from photo-oxidation of oil spill residues. They estimated that about half of the surface oil floating on the GOM in the aftermath of the 2010 Deepwater Horizon spill should have transformed into OxHCs within days to weeks. However, since OxHCs are complex compounds that are outside the analytical window of traditional gas chromatography based techniques, they pose considerable challenges for assessing the environmental risks. Aeppli *et al.*'s study has also identified a suite of oxygenated aliphatic compounds that are more water-soluble and less hydrophobic than their presumed precursors [20]. Their dissolution experiments showed that chemicals in the OxHC fraction can leach into the water and could potentially pose enhanced ecological risks.

Evans *et al.* [21] collected samples from two distinct beach locations in Grand Isle, Louisiana, that were impacted by the DWH oil spill. One of these beaches had higher wave energy than the other. They analyzed the samples for various petroleum contaminants, including PAHs, and concluded that the sample collected from higher energy locations showed a higher degree of weathering. Turner *et al.* [22•] collected DWH oil spill samples from Louisiana marshes for over eight years. They reported that the hydrocarbon levels in the marshes are about 10 times higher than pre-spill conditions. In the case of sediments, although the aerobic sediments returned to pre-spill conditions, the anaerobic sediments appear to retain higher molecular weight hydrocarbons that can adversely affect the health of coastal ecosystems. Karthikeyan *et al.* [23••] subjected oil-contaminated sediments to alternating oxic and anoxic conditions in a laboratory experiment. They reported that under oxic conditions there was a fivefold decrease in total petroleum hydrocarbons compared to that of anoxic conditions, providing further evidence that oxic environments promote petroleum hydrocarbon degradation compared to anoxic environments. Bociu *et al.* [24••] quantified the degradation levels of DWH oil spill residues buried in the upper 50 cm of a sandy beach in Pensacola, Florida. The time-series data for hydrocarbon mass, carbon content, and concentrations of n-alkanes and PAHs indicated very slow degradation. They estimated that the decomposition of DWH oil spill residues embedded in beach sand would take at least 32 years, while degradation without sediment contact may require more than 100 years.

Toxicity and environmental impacts of DWH oil spill residues

Studies have shown that the PAHs present in weathered DWH oil spill residues can be high and they can pose a considerable risk to both human and ecological systems. Brown-Peterson *et al.* [25] exposed juvenile southern flounder to DWH oil-contaminated sediments to assess their impacts on a commercially important benthic fish. Their results show that the exposed flounder length and weight were lower compared to controls after 28 days of exposure. Histopathological analyses showed an increased occurrence of gill abnormalities, including telangiectasis, epithelial proliferation, and fused lamellae in flounders that were exposed to sediments with high PAH concentration. Esbaugh *et al.* [26] completed a mahi-mahi spawning study to assess the effect of embryonic exposure to DWH oil. The study found that the exposure resulted in cardiotoxicity, which was evident from pericardial edema and reduced atrial contractility. This sublethal cardiotoxicity effect could affect the long-term survival of this fish species. Boulais *et al.* [27] evaluated the sublethal effects of sediments contaminated with

DWH oil on gametes, embryos, and veliger larvae of Eastern oysters. Their data show that the contaminated sediments inhibited fertilization. Embryo exposure resulted in developing various abnormalities and reductions in shell growth. Fertilization success and abnormality of larvae exposed as embryos were the most sensitive endpoints for assessing the toxicity effect.

Xu *et al.* [28] exposed water accommodated fractions of weathered DWH crude oil to larval red drum, an estuarine fish species. Their data show that the DWH oil had a significant sublethal effect; the impacts ranged from impaired nervous system to abnormal cardiac morphology. They also reported differentially expressed transcripts, enriched gene ontology, and altered canonical pathways, which will lead to adverse outcomes in nervous and cardiovascular systems. Magnuson *et al.* [29] exposed embryonic zebrafish larvae (4 hour post-fertilization) to weathered oil and evaluated changes in visual functions by tracking optokinetic response. They also assessed cardiotoxicity effects by measuring the heart rate, stroke volume, and cardiac output. Their results show that the zebrafish larvae exposed to crude oil exhibited an increased occurrence of bradycardia. Also, the genes important in eye development and phototransduction were downregulated in oil-exposed larvae, with an increased occurrence of cellular apoptosis, reduced neuronal connection, and reduced optokinetic behavioral response.

Ramesh *et al.* [30] evaluated the changes in the behavioral parameters, hematological markers, liver, and kidney functions in rodents that were exposed to DWH oil. C57 Bl6 mice were exposed to DWH oil and/or Corexit-9500A dispersant. Their study demonstrated that both DWH oil and Corexit-9500A altered the white blood cells and platelet counts. The contaminants also affected the lipid profile and induced toxic effects on the liver and kidney functions. Bhattacharya *et al.* [31] studied the neurotoxic effects of DWH tarmat residues. The water accommodated fraction (WAF) of tarmat was used to quantify the cytotoxicity effects by using MTT assays and cellular morphology assessment. Markers of oxidative stress and apoptosis were assessed to quantify the toxicity effects. They found the tarmat WAF induced a dose-dependent cellular toxicity effect. The toxic chemicals trapped in the tarmat inhibited the cell viability in the hippocampal (H19), kidney (HEK-293), and epithelial (MCF-10A) cells.

Discussions, lessons learned, and some unanswered questions

The published studies related to the DWH oil spill event provide conflicting conceptual paradigms regarding the long-term fate of the toxic compounds present in oil spill residues. For example, during the early days of the spill, researchers have commonly assumed that GOM has highly efficient oil-degrading microorganisms that can rapidly degrade the spilled oil. About a month after shutting down the leaking well, an interagency study completed by the Department of the Interior and the National Oceanic and Atmospheric Administration (NOAA) concluded that out of the 4.9 million barrels of oil spilled, 75% had been cleaned up either by human aid or by naturally occurring environmental processes [32]. This article cited a marine scientist who stated: "*the message I've heard is that everywhere we look, oil is degrading extremely rapidly*" [32]. This paradigm was partially developed based on the data reported in *Science* that attempted to quantify intrinsic bioremediation rates and concluded: "*despite the varying field and microcosm conditions, the oil half-lives are 1.2 to 6.1 days*" [33]. Later, Edwards *et al.* [34] investigated the microbial respiration of offshore surface water and reported that the indigenous microbial community in the GOM possesses the potential to rapidly degrade the spilled oil. These studies have led to the development of an optimistic outlook that the DWH oil spill residues should degrade rather rapidly (within in months or a few years). However, some of the field observations made at other historic oil spill sites, such as the Exxon Valdez spill [35] and the 1991 Gulf War oil spill [36•], have indicated that oil spill residues could persist in the environment for several decades.

NOAA scientists who have followed the Exxon Valdez spill for a long time recently made the following statement [37]: "*the early years after the (Exxon Valdez oil) spill, experts anticipated that the oil would naturally degrade and not persist in the environment. After repeated visits to specific sites over the last 15 years, I haven't found this to be the case. For these sites, the oil may be in the environment for a long time*". A field study completed by this NOAA team also pointed out that the Exxon Valdez oil spill cleanup efforts were terminated after about three years, and the experts predicted that the residual oil would continue to weather rapidly and dissipate within a short time scale. However, contrary to these early optimistic projections, the follow-up field studies have shown that some of the residual oil trapped in beaches continues to persist [38]. Based on a detailed investigation completed between 2001–2015, they concluded that there was very little evidence for change in oil area or mass over the 14 year period.

A few months after the 1991 Gulf War oil spill, a study published in *Nature* concluded that the severe oil pollution was restricted primarily to the Saudi Arabian coastline within about 400 km from the spill; and they also reported that within the first four months the spilled oil has extensively degraded [39]. However, more recently, Arekhi *et al.* [36•] completed a field study along the Northern Qatar shoreline, about 600 km away from Kuwait, and found extensive buried oil spill residues that were formed from the Kuwaiti oil spill. Laboratory analysis of the samples collected from these field sites indicated that the tarmat samples collected in 2019 closely matched the Kuwaiti and Basrah crude oil chemical fingerprints, indicating that these residues must have originated from the Gulf War oil spill. The study also reported that these 27-year old tarmats contain high concentrations of toxic PAHs such as chrysene and its alkylated homologs. These results show that several higher molecular weight PAHs trapped in these buried Kuwait oil spill residues have not degraded even after about three decades of natural weathering.

During the initial days of the DWH oil spill accident, even when the well was actively spewing about 50 000 barrels of oil per day into the GOM, a few researchers boldly predicted that natural processes, that is, 'Mother Nature,' can clean the spill [1]. Our reliance on the ability of Mother Nature to solve anthropogenic environmental contamination problems is not new. For example, civilizations have depended on natural rivers to clean sewage wastes. This approach worked well until we started to discharge large volumes of untreated sewage directly into rivers at rates well above the assimilative capacity of these rivers. The practice of depending on Mother Nature has eventually destroyed the ecological health of many river basins worldwide. We have now understood that rivers have a finite capacity to assimilate wastes. While the GOM is a vast water body, it also has a finite capacity to assimilate wastes at a finite rate. Large oil spills have the potential to exhaust the assimilative capacity of enclosed water bodies such as GOM.

When we depend on Mother Nature for managing large-scale oil spills, we conceptually assume a combination of five major weathering processes, which include biodegradation, photo-oxidation, evaporation, dissolution/dispersion and sedimentation, to remediate the spill. We typically speculate that these five processes will synergistically work together and remove all the toxic compounds in the spilled oil. However, this speculation could largely be a fallacy. If we closely examined these five processes, the last three are physical processes that will not degrade the toxic waste into innocuous products. They are simply phase-transformation processes that primarily depend on dilution to manage (or hide) the problem, with a hope that the diluted contaminants will later be biodegraded by the natural system. Photo-oxidation is a chemical process that can transform contaminants. In recent years, researchers have identified photo-degradation as an important removal mechanism [19,40••]. However, caution should be exercised since photo-oxidation of crude oil contaminants can form oxygenated by-products that are difficult to quantify using standard analytical methods [20]. The OxHCs can persist in the weathered oil/sand tarballs and act as a long-term source of dissolved contaminants. Aquatic organisms could, therefore, be exposed to these oxygenated petroleum products for a long period. It is currently not known whether such exposure could lead to any long-term toxic effects [20].

Based on our current understanding, biodegradation appears to be the only useful process that has the potential to safely mineralize toxic petroleum compounds into harmless (or less harmful) products. However, the availability of nutrients and oxygen could be the limiting factors that might severely limit the biodegradation rates of emulsified oil trapped in buried tarballs or tarmats. Thus far none of the tarball studies has shown the potential of microbes to degrade heavy toxic compounds, such as higher molecular weight PAHs, at a reasonable rate. Most studies tend to report oil bioremediation rate, which is a lumped parameter that characterizes the removal rate of multiple petroleum compounds in floating oil through various processes including dissolution, dispersion, evaporation, and sedimentation. As per our knowledge, no one has completed careful laboratory studies using DWH residues to quantify the degradation rates of specific toxic compounds using controlled experiments that can track the mass balances of various biodegradation by-products. The field study by Bociu *et al.* [24••] is perhaps the only research that used actual tarball residues to conduct field-scale experiments with some level of control. They concluded that the DWH oil spill residues could take up to 30–100 years to degrade. These findings are consistent with an Exxon Valdez study which showed that the oil trapped in the beach environment has not weathered after 25 years of natural weathering. Based on these observations, one could safely conclude that we tend to overly rely on Mother Nature to solve oil spill problems. Caution should be exercised since our knowledge of Mother Nature is limited. Even after a large research investment to study the DWH oil spill for over ten years, we still cannot answer several basic questions related to the spill such as: what are the toxic compounds in the DWH oil that can be biodegraded by the GOM microbes? What are the rates of biodegradation for various petroleum species? What are the biodegradation pathways?

Despite all these uncertainties, the early efforts to clean DWH tarballs deposited along GOM beaches ended on June 10th, 2013. After officially ending the cleanup operations, the Coast Guard released this optimistic press statement: "*more than three years after the worst oil spill in U. S. history erupted in the Gulf of Mexico, the coastlines of Mississippi, Alabama, and Florida have been returned to as close to pre-spill conditions as possible*" [41]. Studies completed over the past 10 years show that the DWH oil spill has indeed substantially increased the background tarball contamination level in Alabama beaches [2,6••,14]. It is currently unclear how long it would take for these beaches to recover to pre-oil-spill conditions, and what will be the long-term impacts. Detailed assessment using an integrated oil spill risk assessment framework (e.g.

Amir-Heidari *et al.* [42]) is needed to quantify the long-term environmental risks posed by these tarballs. Also, while the beaches are indeed recovering with time, as indicated by the reductions in the tarball deposition levels and also the reduction in the average size of the tarballs, it is unclear whether these tarballs are degrading to a non-toxic material. The reductions observed could simply be due to physical deterioration and dilution, wherein the tarballs are broken into smaller fragments and mixed with coastal sediments to a point where they cannot be detected by the naked eye.

Finally, the field observations made over the past 10 years question the conventional paradigm that alleges that the oil spills are expected to yield highly weathered, floating tarballs after years of ocean-scale weathering. It is interesting to note that although over 785 million liters of crude was discharged into the GOM, there is currently no documented evidence for the formation of highly weathered, black, floating, sand-free, traditional-looking tarballs that originated for the DWH oil. All the DWH tarballs recovered from GOM beaches are sunken oil–sand mixtures, which were formed rapidly within days after the oil started to wash along the GOM beaches. What happened to the DWH oil that was stranded and continued to float on the ocean? Will they ever form highly weathered, floating DWH tarballs? These and several other questions related to the long-term environmental impacts of DWH oil spill residues continue to remain unanswered.

Conflict of interest statement

Nothing declared.

CRediT authorship contribution statement

T Prabhakar Clement: Conceptualization, Methodology, Investigation, Writing – original draft, Writing – review & editing, Visualization, Supervision, Resources, Project administration, Funding acquisition. **Gerald F John:** Methodology, Investigation, Writing – original draft, Writing – review & editing, Formal analysis.

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